Non-exponential relaxation in disordered complex systems

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(Dated: February 6, 2008)

We have analytically obtained the non-exponential relaxation function for disordered complex systems applying the multi-level jumping formalism to the fluctuation quantity which makes diffusive motion stochastically in the disordered complex space. It is shown that the relaxation function of disordered complex systems decays obey to stretched exponential law.

PACS numbers: 05.40.-a, 02.50.Ga, 89.75.-k, 77.22.Gm, 61.43.-j

Non-exponential relaxation in disordered complex systems is the object of active research due to its implications in the technology and in several fields of scientific knowledge. The theoretical and experimental studies (for a recent review, see Ref.[1]) show to us that the relaxation behavior in the disordered complex systems such in glasses an supercooled liquids, liquid crystal polymers, dielectrics, magnetic systems, amorphous semiconductors, pinned density wave, protein dynamics, protein folding, and population dynamics among others deviate considerably from the exponential Debye pattern [2]

$$\Phi(t) = \Phi_0 \exp\left[-t/\tau\right]. \tag{1}$$

and it obey to the stretched exponential relaxation function experimentally

$$\Phi(t) = \Phi_0 \exp\left[-t/\tau\right]^{\alpha}, \qquad 0 < \alpha < 1 \tag{2}$$

often referred to as the Kohlrausch-William-Watts (KWW) function [3, 4].

The important task in the relaxation in disordered complex systems is to extend the Debye theory of relaxation of polar molecules to fractional dynamics, so that empirical decay functions, e.g., the stretched exponential of KWW, may be justified. In order to explain the origin of the non-exponential relaxation in the disordered complex systems several models, for example, continuous-time-random-walk models (CTRW) [5, 6, 7, 8, 9, 10, 11], random site energy models [12, 13, 14], defect models [15, 16, 17, 18, 19, 20], jump relaxation models [21], random barrier models [22, 23, 24, 25], hierarchical models [26, 27, 28, 29] have been used and many generalizations [30, 31, 32, 33, 34, 35, 36, 37, 38, 39] of the Debye theory have been suggested up to now.

We remark that one of the most plausible model in among others is definitely phase-space model in which the possible origin of non-exponential relaxation is explained based on the idea of the energy landscape and nontrivial energy barrier so that these models provide a direct link between the phase-space dynamics and slow relaxation. Indeed, several experiments and computer simulations have been done which support the explanation of these relaxation phenomena in the framework of energy landscape paradigm as the result of activated diffusion through a rough energy landscape of valleys and peaks [40, 41]. Therefore, it is suggested that such energetic disorder which produces obstacles or traps which delay the motion of the particle and introduce memory effects into the motion [18]. Hence, in such scenario a process can be characterized by the temporally nonlocal behavior [9].

Considering stochastic framework, in a relaxation process, the fluctuation variable x which may represent the physical quantity such as dipole, jumps at random from one value to another with equal probability and it takes stochastic values as x_1, x_2, x_3,...,x_N with time. Such a stochastic process can be regarded as multi-level jumping process. In this study, using multi-level jumping formalism we shall obtain relaxation function of disordered complex system in which Brownian quantity diffusive as stochastically. The relaxation function for a stochastic process is given simply

$$\Phi(t) = \Psi(t=\infty) - \Psi(t) \tag{3}$$

where $\Psi(t)$ is the response function. The response of the any system can be written in terms of the correlation function as

$$\Psi(t) = \frac{1}{kT}\left(\left\langle x^2\right\rangle - \left\langle x(0)\, x(t)\right\rangle\right) \tag{4}$$

where k is the Boltzmann constant, T is the temperature, and $\left\langle x^2\right\rangle$ is the mean-square average of the x. On the other hand, $\left\langle x(0)\, x(t)\right\rangle$ is the correlation function of the fluctuation variable x. The correlation function can be given in discrete form follows

$$\left\langle x(0)\, x(t)\right\rangle = \sum_{x_0}\sum_{x} p(x_0)\, x_0 P(x_0 \mid x, t)\, x \tag{5}$$

where the quantities x_0 and x are the values of stochastic variable x at times 0 and t, respectively. Having started at the initial state x_0 with the statistical weight $p(x_0)$, $P(x_0 \mid x, t)$ measures the probability of propagation from x_0 to x in time t, which is called as conditional probability. The correlation function measures the decay of the fluctuation variable of the physical quantity in the system, and it is determined due to conditional probability

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which depends on the nature of the stochastic system i.e, the system is whether ordered or disordered complex.

Eqs. (3-5) clearly indicate that relaxation function can be obtained from fluctuation-dissipation theory in the framework linear-response theory if correlation function belong to the relaxation process is known.

The conditional probability in Eq. (5) for disordered complex space may be written formally [43], but suggestive notation, as

$$P\left(x_0 \mid x,t\right) = \delta_{x0} + \frac{1}{\Gamma\left(\alpha\right)} \int_0^t \left(t-t'\right)^{\alpha-1} \times \sum_{x'} W\left(x' \mid x\right) P\left(x_0 \mid x',t'\right) dt' \tag{6}$$

where α is the fractional order which plays the role of a dynamical exponent, δ_{x0} is the initial condition at $t=0$, and $W\left(x \mid x'\right)$ is the jump rate for a particle from x to x'. The Eq. (6) is known the fractal time master equation which defines non-Markovian stochastic process with a memory, which is a special case of the CTRW equation [42, 43].

In the operator formalism [44, 45, 46] the conditional probability $P\left(x_0 \mid x,t\right)$ is simply given with the matrix element of operator $\widehat{P}(t)$ as

$$P\left(x_0 \mid x,t\right) \equiv \left\langle x\right| \widehat{P}\left(t\right) \left|x_0\right\rangle . \tag{7}$$

where the conditional probability satisfy

$$P\left(x_0 \mid x,0\right) \equiv \left\langle x\right| \widehat{P}\left(0\right) \left|x_0\right\rangle = \delta\left(x-x_0\right) \tag{8}$$

for $t=0$. Similarly, $\widehat{W}$ is also the jump operator which is given by

$$W\left(x' \mid x\right) = \left\langle x\right| \widehat{W} \left|x'\right\rangle . \tag{9}$$

Matrix representation allows to us to write the physical quantities as an operator. Hence, the correlation function in Eq. (5) can be transformed to the matrix representation

$$\left\langle x\left(0\right) x\left(t\right)\right\rangle = \sum_x p(x_0) \left\langle x_0\right| \widehat{X} \left|x_0\right\rangle \left\langle x\right| \widehat{P}\left(t\right) \left|x_0\right\rangle \left\langle x\right| \widehat{X} \left|x\right\rangle \tag{10}$$

where $\widehat{X}$ is the matrix representation of the fluctuation quantity x, $\widehat{P}(t)$ is the matrix form of the conditional probability, and $\left|...\right\rangle$ indicates vector space of stochastic states.

The fact that x takes N valued stochastic variable in which the stochastic states have been represented in N dimensional vector space. Accordingly, the fluctuation variable x and conditional probability $P\left(x_0 \mid x,t\right)$ may be given by the matrix representation associate a stochastic state $\left|x\right\rangle$ ($x = 1,2,3,...,N$ for multi-level jumping process) with N values [46]

$$\left|1\right\rangle = \begin{pmatrix} 1 \\ 0 \\ 0 \\ . \\ . \\ . \\ 0 \end{pmatrix}, \left|2\right\rangle = \begin{pmatrix} 0 \\ 1 \\ 0 \\ . \\ . \\ . \\ 0 \end{pmatrix}, ..., \left|N\right\rangle = \begin{pmatrix} 0 \\ 0 \\ 0 \\ . \\ . \\ . \\ 1 \end{pmatrix} \tag{11}$$

The stochastic states $\left|x\right\rangle$ (and similarly $\left|x'\right\rangle$) is taken to form an orthonormal set which provide *closure* property as

$$\sum_x \left|x\right\rangle \left\langle x\right| = 1 \tag{12}$$

for discrete variables, and assign to it an *apriori* occupation probability $p(x) = \frac{1}{N}$.

Hence, the fluctuation variable x can be also given by matrix form, the eigenvalues of which correspond to stochastic variables x_1, x_2, x_3,...,x_N, respectively. The matrix form of the operator $\widehat{X}$ of the fluctuation variable x is represented as

$$\widehat{X} = \begin{pmatrix} x_1 & 0 & 0 & . & . & . & 0 \\ 0 & x_2 & 0 & . & . & . & 0 \\ 0 & 0 & x_3 & . & . & . & 0 \\ . & . & . & . & . & . & . \\ . & . & . & . & . & . & . \\ . & . & . & . & . & . & . \\ 0 & 0 & 0 & . & . & . & x_N \end{pmatrix} \tag{13}$$

Now, after these definitions, the correlation function Eq. (6) can be carried out using this formalism, however, we need the operator form of the conditional probability in order that calculation of it.

The time derivative of Eq. (5) is written as

$$\frac{\partial}{\partial t} P\left(x_0 \mid x,t\right) = \frac{1}{\Gamma\left(\alpha\right)} \frac{\partial}{\partial t} \int_0^t \left(t-t'\right)^{\alpha-1} \sum_{x'} W\left(x' \mid x\right) P\left(x_0 \mid x',t'\right) dt' \tag{14}$$

It is seen that Eq. (14) contains a convolution integral with a slowly decaying power-law Kernel $M\left(t\right) = t^{\alpha-1}/\Gamma\left(\alpha\right)$, ensures the non-Markovian nature of the subdiffusion process defined by the fractional diffusion process. This convolution integral is defined as an operator which is known as fractional Riemann-Liouville integro-differential operator [47];

$${}_0D_t^{1-\alpha} P\left(x_0 \mid x,t\right) = \frac{1}{\Gamma\left(\alpha\right)} \frac{\partial}{\partial t} \int_0^t dt' \frac{P\left(x_0 \mid x',t'\right)}{\left(t-t'\right)^{1-\alpha}} \tag{15}$$

with $0 < \alpha < 1$.

Eq. (14) can be reduced to simply form as down

$$\frac{\partial}{\partial t} P\left(x_0 \mid x,t\right) ={}_0 D_t^{1-\alpha} \sum_{x'} W\left(x' \mid x\right) P\left(x_0 \mid x',t'\right) \tag{16}$$

using Eq. (15). On the other hand, using operator formalism in Eqs. (7) and (9), Eq. (16) can be rewritten completely matrix form

$$\frac{\partial}{\partial t}\left\langle x\right|\widehat{P}\left(t\right)\left|x_0\right\rangle ={}_0 D_t^{1-\alpha}\sum_{x'}\left\langle x\right|\widehat{W}\left|x'\right\rangle\left\langle x'\right|\widehat{P}\left(t\right)\left|x_0\right\rangle . \tag{17}$$

Using closure property in Eq. (12), Eq. (17) is reduced to

$$\left\langle x\right|\frac{\partial\widehat{P}\left(t\right)}{\partial t}\left|x_0\right\rangle ={}_0 D_t^{1-\alpha}\left\langle x\right|\widehat{W}\widehat{P}\left(t\right)\left|x_0\right\rangle . \tag{18}$$

This equation can be simplify as

$$\frac{\partial}{\partial t}\widehat{P}\left(t\right)={}_0 D_t^{1-\alpha}\widehat{W}\widehat{P}\left(t\right). \tag{19}$$

Eq. (19) is known fractional relaxation equation [9, 48]. The solution of it may be presented in terms of Mittag-Leffler function [49]

$$\widehat{P}\left(t\right)=E_\alpha\left[\widehat{W}t^\alpha\right]=\sum_{j=0}^{\infty}\frac{\left(\widehat{W}t^\alpha\right)^j}{\Gamma\left(1+\alpha j\right)}. \tag{20}$$

For $\alpha = 1$ the Mittag-Leffler function has the standard exponential form

$$E_{\alpha=1}\left[\widehat{W}t\right]=\exp\left[\widehat{W}t\right] \tag{21}$$

whereas for $0 < \alpha < 1$ it interpolates the initial stretched exponential form as

$$E_\alpha\left[\widehat{W}t^\alpha\right]\backsim\exp\left[\frac{\widehat{W}t^\alpha}{\Gamma\left(1+\alpha\right)}\right] \tag{22}$$

and, however, at the long time, the the initial stretched exponential behavior turns over to the power-law behavior

$$E_\alpha\left[\widehat{W}t^\alpha\right]\backsim\frac{1}{\Gamma\left(1+\alpha\right)\widehat{W}}t^{-\alpha}. \tag{23}$$

In this study, we have focused the behavior of short time limit, and we have written the conditional probability (20) for the the simplicity as down

$$\widehat{P}\left(t\right)\equiv\exp\left[\widehat{W}t^\alpha\right] \tag{24}$$

using Eq. (22) form. Actually, this result clearly indicates that the character of the relaxation has stretched exponential i.e. KWW form for the disordered complex systems. The jump matrix $\widehat{W}$ in Eq. (24) may be represented in terms of collision and unit matrix elements as

$$\widehat{W}=\lambda\left(\widehat{J}-\mathbf{1}\right) \tag{25}$$

where λ is the relaxation rate, $\widehat{J}$ is the collision matrix, and $\mathbf{1}$ is a unit matrix.

For the N-level the relaxation rate is given by

$$\lambda = Nw. \tag{26}$$

and the collision matrix $\widehat{J}$ is prescribed by

$$\widehat{J}=\begin{pmatrix} 1/N & 1/N & . & . & . & 1/N \\ 1/N & 1/N & . & . & . & 1/N \\ . & . & . & . & . & . \\ . & . & . & . & . & . \\ . & . & . & . & . & . \\ 1/N & 1/N & . & . & . & 1/N \end{pmatrix} \tag{27}$$

where w is the jump rate from one value of x to another.

The jump matrix $\widehat{W}$ is also given by

$$\widehat{W}=\begin{pmatrix} (1-N)w & w & . & . & . & w \\ w & (1-N)w & . & . & . & w \\ . & . & . & . & . & . \\ . & . & . & . & . & . \\ . & . & . & . & . & . \\ w & w & . & . & . & (1-N)w \end{pmatrix} \tag{28}$$

The usefulness of the decomposition in Eq. (25) is borne out by the fact that the $\widehat{J}$ matrix has a very simple property; it is *idempotent*, i.e.,

$$\widehat{J^2}=\widehat{J},\quad \widehat{J^3}=\widehat{J},\quad ...,\quad \widehat{J^k}=\widehat{J} \tag{29}$$

for any integer $k > 0$. This property allows us to immediately construct the conditional probability $\widehat{P}(t)$ in matrix form:

$$\widehat{P}\left(t\right)=\exp\left[\lambda\left(\widehat{J}-\mathbf{1}\right)t^\alpha\right] \tag{30}$$

Hence, using direct power series expansion and Eq. (29), Eq. (30) can be written follow

$$\widehat{P}\left(t\right)=\exp\left(-\lambda t^\alpha\right)\left[\mathbf{1}-\widehat{J}+\widehat{J}\exp\left(\lambda t^\alpha\right)\right]. \tag{31}$$

It is convenient to introduce the more general notation for basis vectors in order to make easy of calculations. Therefore, let us associate a stochastic state $\left|n\right\rangle$ ($n = 1, 2, 3, ..., N$ for multi-level jumping process) instead of $\left|x\right\rangle$. Thus, it is possible that Eq. (10) is rewritten down form

$$\left\langle x\left(0\right)x\left(t\right)\right\rangle=\sum_{n,m}p_n\left\langle n\right|\widehat{X}\left|n\right\rangle\left\langle m\right|\widehat{P}\left(t\right)\left|n\right\rangle\left\langle m\right|\widehat{X}\left|m\right\rangle \tag{32}$$

with *apriori* occupation probability

$$p_n(x)=\frac{1}{N},\qquad n=1,2,...,N. \tag{33}$$

Now, it is possible expectation values of the quantities which we interested in;

$$\left\langle n\right|\widehat{P}\left(t\right)\left|m\right\rangle=\frac{1}{N}+\left(\delta_{nm}-\frac{1}{N}\right)\exp\left(-\lambda t^\alpha\right) \tag{34}$$

where $n, m = 1, 2, ..., N$. The matrix of the operator $\widehat{X}$ is

$$\langle n| \widehat{X} |m\rangle = X_n \delta nm, \qquad n, m = 1, 2, ..., N \tag{35}$$

where X_n are the allowed values of the stochastic variables. In addition, average of the collision matrix $\widehat{J}$ is presented as

$$\langle n| \widehat{J} |m\rangle = \frac{1}{N}, \qquad n, m = 1, 2, ..., N \tag{36}$$

If Eqs. (34) and (35) are inserted in Eq. (10), after a bit of algebra, the correlation function can be written down form

$$\langle x(0)\, x(t)\rangle = \langle x\rangle^2 + \left(\langle x^2\rangle - \langle x\rangle^2\right) \exp\left(-\lambda t^{\alpha}\right) \tag{37}$$

where the deterministic quantities are weighted averages over the available states of the corresponding variable. The average value of $\langle x\rangle$ and $\langle x^2\rangle$ in the stationary state are given by

$$\langle x\rangle = \sum_{n=1}^{N} p_n \langle n| \widehat{X} |n\rangle = \frac{1}{N} \sum_{n=1}^{N} X_n \tag{38}$$

and

$$\langle x^2\rangle = \sum_{n=1}^{N} p_n \langle n| \widehat{X^2} |n\rangle = \frac{1}{N} \sum_{n=1}^{N} X_n^2 \tag{39}$$

respectively.

The response function $\Psi(t)$ in Eq. (4) for this process is obtained as

$$\Psi(t) = \frac{1}{kT} \left\{ \langle x^2\rangle - \langle x\rangle^2 - \left[\langle x^2\rangle - \langle x\rangle^2\right] \exp\left[-\lambda t^{\alpha}\right] \right\} \tag{40}$$

using Eq. (37), on the other hand, the limit the behavior of $\Psi(t)$ at the infinity is presented as

$$\Psi(t = \infty) = \frac{1}{kT} \left(\langle x^2\rangle - \langle x\rangle^2\right). \tag{41}$$

When Eqs. (40) and (41) are inserted into Eq. (3), the relaxation function can be obtained down form

$$\Phi(t) = \frac{1}{kT} \left\langle (\Delta x(t))^2 \right\rangle \exp\left[-\lambda t^{\alpha}\right] \tag{42}$$

where $\left\langle (\Delta x)^2 \right\rangle$ is the mean squared displacement which contains solely the molecular contributions

$$\left\langle (\Delta x(t))^2 \right\rangle = \langle x^2\rangle - \langle x\rangle^2 = \frac{2K_{\alpha}}{\Gamma(1+\alpha)} t^{\alpha} \tag{43}$$

where K_{α} is known the diffusion constants which is a generalization of the Einstein-Stokes-Smoluchowski relation [9]. The K_{α} is defined as

$$K_{\alpha} = k_B T / m\eta_{\alpha} \tag{44}$$

where η_{α} is the friction coefficient which is a measure for interaction of the particle with its environment, and m denotes mass of the particle [9, 50, 51, 52].

Finally the relaxation function in Eq. (42) can be simplified as

$$\Phi(t) = \frac{1}{kT} \frac{2K_{\alpha}}{\Gamma(1+\alpha)} t^{\alpha} \exp\left[-\lambda t^{\alpha}\right]. \tag{45}$$

The prefactor in Eq. (45) can be taken Φ_0 which is a constant in Eq. (2). As a result, the relaxation function (45) is consistent with Eq. (2), which clearly indicates that fluctuation quantity in a disordered complex system decays with time as stretched exponential i.e, KWW form.

In this study, applying the multi-level jumping formalism to the fluctuation quantity which make diffusive motion stochastically in the disordered complex system, we have analytically obtained the relaxation function KWW form in terms of correlation function in absence of the external field. It is concluded that multi-level jumping process formalism is quite powerful technique for the modelling of the Brownian motion to obtain the relaxation function of disordered complex systems.

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